Correlations and Néel Order of Randomly Diluted Quantum Spin Ladders

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We present a Monte Carlo study of the correlation length ξ of randomly diluted antiferromagnetic Heisenberg ladders, composed of two spin–1/2 chains. For weak and intermediate inter–chain couplings, $J_\perp/J \leq 1$, we find an enhancement of correlations that is strongest for a fraction $z^* \approx J_\perp/(8J)$ of dilutants. We are able to access the experimentally relevant low–temperature regime, $T/J \approx 1/500$, and find that the recently inferred Néel temperature of $Sr(Cu_{1-z}Zn_z)_2O_3$ corresponds to a curve of constant correlation length $\xi \approx 18$ of the single diluted ladder with $J_\perp/J \lesssim 1/2$. The primary reason for the Néel ordering is argued to be a strong enhancement of two–dimensional correlations due to a Cu–Sr–Cu exchange coupling of $\approx 10 meV$ in the stacking direction of the ladders.

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Low–dimensional quantum Heisenberg antiferromagnets exhibit many unusual collective properties owing to the increased importance of quantum fluctuations when the dimensionality of the system is less than three, and to the continuous $O(3)$ spin rotational symmetry of Heisenberg interactions. The one–dimensional (1D) Heisenberg chain and its two–dimensional (2D) analog, the square–lattice antiferromagnet, have been studied extensively for many decades. Arguably, the most prominent examples in this context are the lamellar copper oxides, which in their undoped states are square–lattice Heisenberg antiferromagnets with predominant nearest–neighbor (NN) interactions and a spin value of $S = 1/2$ [1]. The properties of the NN Heisenberg chain are very different from those of the square lattice. Instead of true long–range order, the $S = 1/2$ chain exhibits a critical ground state characterized by algebraic decay of the spin correlations and gapless excitations [2]. In 1983, Haldane [3] conjectured that the basic properties of the $S = 1/2$ chain are shared only by chains with half–odd–integer spin, and that integer–S chains should behave fundamentally differently. The latter were predicted to possess a spin–liquid ground state characterized by a finite correlation length ξ_0 due to the presence of a gap $\Delta \sim 1/\xi_0$ to the lowest excitations. These predictions have been confirmed both numerically and experimentally [4].

In recent years, the question of how quantum fluctuations manifest themselves in systems whose extent is finite in one dimension and infinite in another has received much attention. In particular, arrays of coupled chains of width n, so–called spin ladders, have been studied extensively [5]. Good physical realizations of these systems, with $S = 1/2$, were found to exist in materials such as $SrCu_2O_3$ ($n = 2$) and $Sr_2Cu_3O_5$ ($n = 3$) [6]. In close analogy to the behavior of the spin–S chains, the properties of NN $S = 1/2$ ladders are fundamentally different for even and odd numbers of coupled chains [5]. Quite generally, it is believed that quantum fluctuations always generate a spin gap when the product nS is an integer,

and that the destructive interference of fluctuations leads to a gapless spectrum for half–odd–integer values of nS [7]. Theory for the temperature dependence of the spin correlations of $S = 1/2$ ladders [8] is in good agreement with Monte Carlo results [8,9].

A surprising recent experimental discovery has been that random dilution in the ladder material $Sr(Cu_{1-z}Zn_z)_2O_3$ ($S = 1/2$, $n = 2$) leads to antiferromagnetic long–range order at low temperatures, even for very small Zn concentrations [10]. A schematic of the structure of this material is shown in Fig. 1(a). The observed Néel order is counter–intuitive, since based on classical intuition one might have expected a diminution of correlations due to dilution, and hence a reduced tendency to order. A large number of numerical and theoretical studies have addressed this issue [11], yet a quantitative understanding of the experimentally relevant low–temperature spin correlations of a diluted two–chain ladder is still lacking. Furthermore, the explicit pathway by which dilution promotes three–dimensional (3D) Néel order remains elusive.

In this paper, we present a numerical investigation of the effects of random dilution z on the spin correlations of a $n = 2$, S = 1/2, NN antiferromagnetic Heisenberg ladder. We are able to reach the experimentally relevant low–temperature regime, $T/J \approx 1/500$, and find that a small amount of dilution indeed enhances the antiferromagnetic correlation length, albeit rather weakly. For weak and intermediate inter–chain couplings, $J_\perp/J \leq 1$, we find that the enhancement is strongest for an average spacing between dilutants that approximately equals $\xi_0 = \xi(z = 0, J_\perp/J, T/J = 0)$, the zero–temperature correlation length of the un–diluted ladder. In the limit of small $J_\perp/J$, we have shown previously that $\xi_0 \approx 3.9J/J_\perp$ [9]. Since the Néel temperature $T_N(z)$ of $Sr(Cu_{1-z}Zn_z)_2O_3$ peaks at $z \approx 4\%$ [10], this in turn requires that $J_\perp/J \lesssim 1/2$. This bound is consistent with recent experimental and theoretical estimates for the ratio $J_\perp/J$. Moreover, we find that $T_N(z)$

of $Sr(Cu_{1-z}Zn_z)_2O_3$ corresponds to a curve of constant correlation length $\xi \approx 18$ of a single ladder. We argue that the primary reason for the experimentally observed divergence of spin correlations at $T_N(z)$ is a crossover from 1D to 2D behavior due to the inter–ladder Cu–Sr–Cu exchange in the stacking direction of the ladders; an important observation is the fact that the Cu–Sr–Cu geometry and distances are very similar to those in the Cu–Y–Cu bi–layer in the high–T_c precursor material $YBa_2Cu_3O_{6+\delta}$. From neutron scattering measurments (for $\delta = 0.15 - 0.20$) it is known that the bi–layer coupling is $\approx 10meV$ [12]. The final 3D antiferromagnetic ordering then occurs because of a very weak pseudo-dipolar coupling between the frustrated ladder segments [13]. We thus predict a broad 1D to 2D crossover of the spin correlations followed by sharp 3D Néel ordering transition. This could be observed directly in neutron scattering experiments once suitably large single crystals become available.

As in our previous studies [8,9], the ladders are investigated with a very efficient loop cluster algorithm [14]. The Hamiltonian operator for a Heisenberg ladder is

$$\mathcal{H} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_\perp \sum_{\langle ij \rangle_\perp} \mathbf{S}_i \cdot \mathbf{S}_j, \qquad (1)$$

where $\mathbf{S}_i = \frac{1}{2}\sigma_i$ is the quantum spin operator at each site i of non–zero spin, while $\langle ij \rangle$ and $\langle ij \rangle_\perp$ denote nearest neighbors along the chains and across the rungs of the ladder, respectively (see Fig. 1(b)). The couplings considered are antiferromagnetic, that is, J and $J_\perp > 0$. Periodic boundary conditions are employed only along the chains, and we use units in which the lattice constant is unity, as well as $\hbar = k_B = g\mu_B = 1$. We typically average over 40 random configurations of each 10^4 updates and measurements, and compute the staggered instantaneous correlation function

$$C(i,j) = \text{sign}(i,j)\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle, \qquad (2)$$

where $\text{sign}(i,j) = 1$ if the spins at i and j are on the same sublattice of the antiferromagnet, and $\text{sign}(i,j) = -1$ otherwise. The correlation length of the diluted ladders, $\xi = \xi(z, J_\perp/J, T/J)$, is extracted at large distances r, $r/\xi \geq 3$, from fits to the symmetrized form $C(r) \sim \cosh[(L/2-r)/\xi]$; the length L of the ladders is kept at least an order of magnitude larger than ξ.

In the present study, we focus on ladders with $1/4 \leq J_\perp/J \leq 1$. The dependence of the correlation length on the degree of dilution is shown in Fig. 2. Here, the temperature is chosen such that $T/J_\perp = 3/100$ is constant. As z is increased from zero, ξ is enhanced, reaches a maximum, and eventually decreases at large z. In a previous study of un–diluted ladders [9] we found that at weak inter–chain coupling the product $\Delta\xi$ exhibits a rather simple scaling behavior in the variable T/Δ. The

weak–coupling regime is characterized by the relationship $\Delta \approx 0.41J_\perp$, and it sets in below $J_\perp/J \approx 1/2$ [15]. This motivated us to re–plot the data of Fig. 2 as $\xi J_\perp/J$ versus $zJ/J_\perp$, as shown in the inset. Indeed, the data for $J_\perp = 1/4$ and $1/3$ collapse to a single curve, while those for $J_\perp \geq 1/2$ deviate slightly from this curve.

The enhancement of correlations appears to be largest for an approximate average spacing between dilutants of $\xi_0 = \xi(0, J_\perp/J, 0)$; specifically we find empirically that the maximum occurs for $z^* = \frac{1}{2}\frac{1}{\xi_0+\xi^*}$, where $\xi^* = \mathcal{O}(1)$ is a constant. The weak–coupling regime is characterized by $\Delta \approx 0.41J_\perp$ and $\xi_0 = c_{1D}/\Delta$, where $c_{1D} = \pi/2$ is the spin–wave velocity of the single spin–1/2 chain [9]. Deep in the weak–coupling regime, $\xi_0 \approx 3.9J/J_\perp$ is large, so that $z^* \approx 0.13J_\perp/J$, consistent with Fig. 2.

For $Sr(Cu_{1-z}Zn_z)_2O_3$, the Néel temperature, and thus the enhancement of correlations, is the largest for $z \approx 4\%$. Based on our observations for the single ladder, this implies that $1/4 \leq J_\perp/J \leq 1/2$. Such a strong exchange anisotropy might at first appear to be somewhat surprising, given that the Cu–O–Cu distances along the chains and across the rungs are nearly identical. However, both recent experiments [16,17] and theoretical calculations [18] imply that $J_\perp/J$ is of order 0.4 to 0.5, consistent with our own deductions.

We now discuss quantitatively the Néel ordering of $Sr(Cu_{1-z}Zn_z)_2O_3$, based on the assumption that $J_\perp/J \approx 1/2$. To effect this, we simulate the diluted ladder with $J_\perp/J = 1/2$ down to very low temperatures, $T/J \approx 1/500$. In our previous study of the the un–diluted ladder ($z = 0$) with $J_\perp/J = 1/2$ we had found that $\xi_0 \approx 7.5$ [9]. The temperature dependences of the correlation length at several non–zero values of z are shown in Fig. 3 together with the previous result for $z = 0$. Corresponding staggered susceptibility data are given in the inset. As noted before, the enhancement of correlations is strongest for $z^* \approx 4\%$.

The maximum ordering temperature of the experimental system is $T_N \approx 8K$ at $z^* \approx 4\%$ [10], as shown in Fig. 4. With $J \approx 1900K$ [16], this corresponds to $T_N/J \approx 0.004$. From Fig. 3, it is apparent that the correlations of the isolated ladder for $z \approx 4\%$ and $T/J \approx 0.004$ are enhanced only to $\xi \approx 18$. In fact, we find that $T_N(z)$ of $Sr(Cu_zZn_{1-z})_2O_3$ corresponds to a curve of constant correlation length, $\xi = 18(1)$, as shown in Fig. 4.

What then is the mechanism that leads to the dramatic enhancement of correlations and the concomitant Néel order observed experimentally? The fact that the experimentally observed $T_N(z)$ corresponds to a curve of constant correlation length of a single diluted ladder implies that this mechanism is independent of the degree of dilution. It is important to realize that the inter–ladder Heisenberg coupling within the Cu–O sheets remains essentially frustrated at non–zero dilution. A key observation in understanding the onset of long–range order in Zn–doped $SrCu_2O_3$ is that the $Cu^{2+} - Sr^{2+} - Cu^{2+}$

geometry (see Fig. 1(a)) and distances in the stacking direction of the ladders are essentially identical to those in the $Cu^{2+} - Y^{3+} - Cu^{2+}$ bi–layer arrangement in $YBa_2Cu_3O_{6+\delta}$; recent neutron scattering experiments, for $\delta = 0.15 - 0.2$, give $J_c \approx 10meV$ [12]. Therefore, the primary perturbation to the quasi–1D ladders in $Sr(Cu_{1-z}Zn_z)_2O_3$ is the coupling between ladders in adjacent planes. The reduced value for this inter–ladder coupling is $\alpha_{2D} = J_c/J \approx 6\%$. Accordingly, we anticipate a progressive crossover from 1D to 2D spin correlations, followed by a sharp transition to 3D Néel order due to a intra–planar pseudo–dipolar coupling between adjacent ladders [13]. This evolution should be observable experimentally once suitably large single crystals become available.

The inset of Fig. 3 shows some results for the staggered susceptibility per spin, $\chi_s \sim T^{-1}\langle(\sum_i(-1)^i S_i^z)^2\rangle$. We find that $J\chi_s \approx 160$ along the curve of constant correlation length $\xi \approx 18$, Fig. 4. Mean–field theory for the enhancement of the susceptibility predicts

$$\chi_s^{MF} = \frac{\chi_s}{1 - J_c\chi_s}, \qquad (3)$$

which diverges for $J\chi_s = 1/\alpha_{2D} \approx 18$. From the inset of Fig. 3 it is clear, that such a small value of $J\chi_s$ is already reached at rather high temperatures. Perhaps not surprisingly, mean–field theory predicts 2D ordering, and, implicitly, 3D ordering, at a temperature which is about an order of magnitude higher than that observed experimentally for all values of z. Empirically, the condition for the Néel ordering appears to be $\xi = 1/\alpha_{2D} \approx 18$, rather than $J\chi_s = 1/\alpha_{2D}$; this remains to be understood theoretically.

A previous numerical study of the susceptibilities of the diluted ladder [19] extended to temperatures as low as $T/J = 1/200$ and used Eq. (3) together with $J_\perp/J = 1$, $J = 1000K$, and $\alpha_{3D} \approx 2\%$, to explain qualitatively the phase diagam of $Sr(Cu_{1-z}Zn_z)_2O_3$. Our results are consistent with this work, but give a physically more complete and precise description.

Recently, Fujiwara et al. [20] presented an interesting NMR study of ^{65}Cu linewidths in $Sr(Cu_{1-z}Zn_z)_2O_3$ at small Zn concentrations, $z \leq 0.5\%$, and and for temperatures $T \geq 20K$. Our results imply that these data cannot be understood with the model suggested by the authors of Ref. [20], since the latter model requires ladder correlation lengths one to two orders of magnitude larger than those which we compute. Further work is required to understand these most interesting NMR results.

In summary, we have numerically determined the correlation length of randomly diluted spin–1/2 two–chain Heisenberg ladders down to the experimentally relevant low–temperature regime, $T/J \approx 1/500$. At weak and intermediate inter–chain couplings, $J_\perp/J \leq 1$, we find an enhancement of correlations that is strongest for a fraction $z^* \approx J_\perp/(8J)$ of dilutants. The recently inferred Néel ordering temperature $T_N(z)$ of $Sr(Cu_{1-z}Zn_z)_2O_3$ is found to correspond well to a curve of constant correlation length $\xi \approx 18$ of the single diluted ladder with $J_\perp/J \lesssim 1/2$. We argue that the primary reason for the experimentally observed 3D Néel ordering is an enhancement of 2D correlations due to an antiferromagnetic coupling $\alpha_{2D} = J_c/J \approx 6\%$ in the stacking direction of the ladders. Empirically, the condition for Néel ordering thus appears to be $\xi = 1/\alpha_{2D}$. The 1D to 2D crossover is then followed by a sharp transition to 3D Néel order due to a tiny intra–planar pseudo–dipolar coupling between adjacent ladders. Once sizable single crystals become available, this predicted evolution of the correlations should be observable in neutron scattering experiments.

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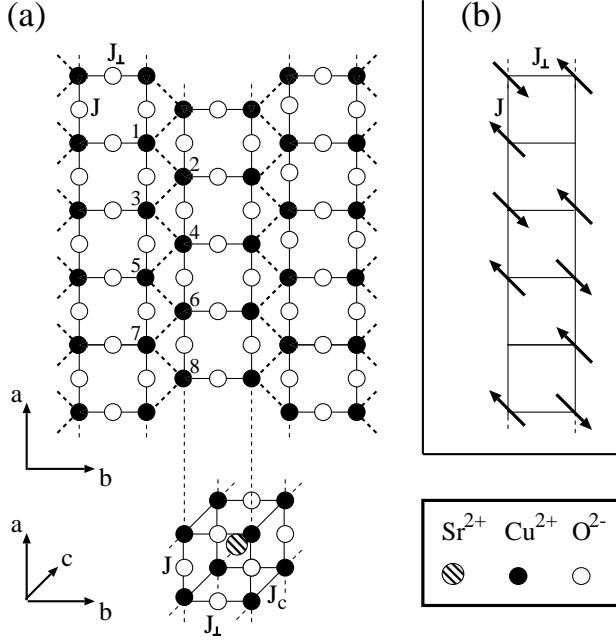[20] N. Fujiwara *et al.*, Phys. Rev. Lett. **80**, 604 (1998).

FIG. 1. (a) Schematic structure of the two–chain ladder compound $SrCu_2O_3$, which has Cu^{2+} spin–1/2 magnetic moments and antiferromagnetic exchange couplings J and $J_\perp$ along the chains and across the rungs, respectively. The isotropic inter–ladder coupling, indicated by the dashed lines, is frustrated; at site 2, for example, the coupling 1 - - 2 is cancelled by the coupling 2 - - 3. J_c is the Cu–Sr–Cu coupling in the stacking direction of the ladders. For $Sr(Cu_{1-z}Zn_z)_2O_3$, a fraction z of the copper spins is randomly replaced by non–magnetic Zn^{2+} ions. (b) Schematic of a single diluted ladder as investigated in the present Monte Carlo study.
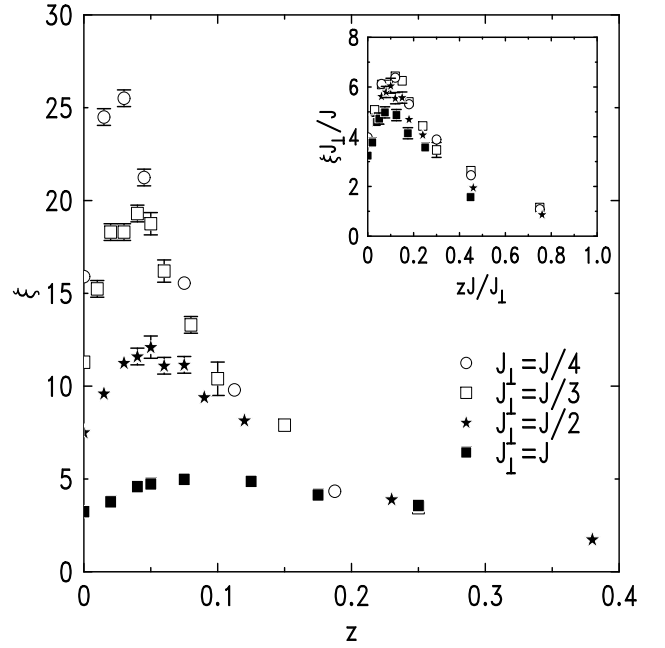

FIG. 2. Correlation length versus degree of dilution of randomly diluted ladders. The temperatures were chosen such that $T/J_\perp = 3/100$. Inset: Evidence for scaling of $\xi J_\perp/J$ in the variable $zJ/J_\perp$.
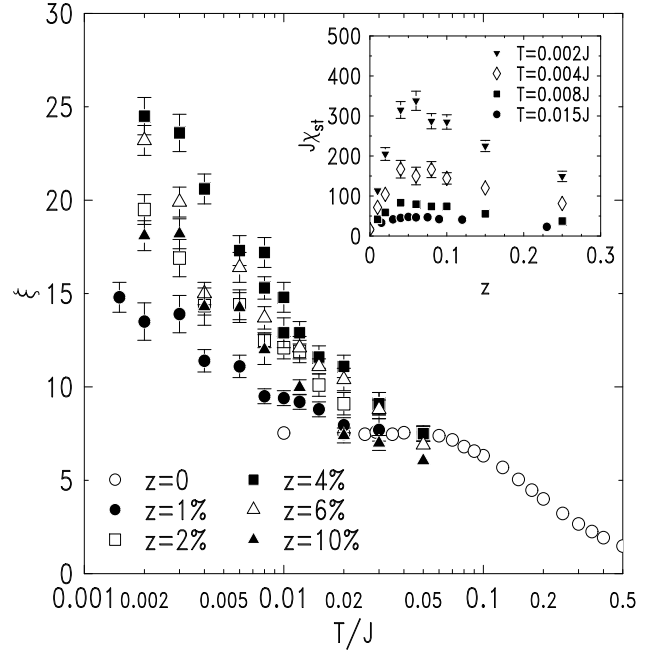

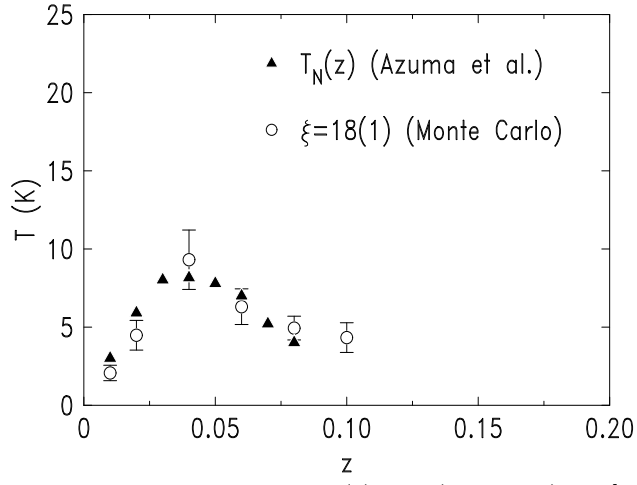FIG. 3. Correlation length and staggered susceptibility (inset) for a randomly diluted ladder with $J_\perp/J = 1/2$.

4

FIG. 4. Néel temperature $T_N(z)$ of $Sr(Cu_{1-z}Zn_z)_2O_3$ [10] and points of constant correlation length $\xi = 18(1)$ of a single diluted ladder with $J_\perp/J = 1/2$, as obtained from Monte Carlo. In this comparison, we use $J = 1900K$ [16].